

Atomic coherence and interference phenomena in resonant nonlinear optical interactions¹

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630090, Novosibirsk, Russia**1 Abstract**

Interference effects in quantum transitions, giving rise to amplification without inversion, optical transparency and to enhancements in nonlinear optical frequency conversions are considered. Review of the relevant early theoretical and experimental results is given. The role of relaxation processes, spontaneous cascade of polarizations, local field effects, Doppler-broadening, as well as specific features of the interference in the spectral continuum are discussed.

Keywords: atomic coherence and interference, resonant nonlinear interactions, bound-free transitions, amplification without inversion, relaxation-induced processes, local field effects, inhomogeneous broadening, frequency-conversion, *VUV* generation

2 Introduction

There has been considerable interest recently in the study of laser-induced quantum coherence and interference, which leads to fundamental effects in high resolution nonlinear spectroscopy, to amplification of radiation without the requirement of population inversion (*AWI*) and to resonantly enhanced refraction at vanishing (without) absorption (*ERWA*), to coherent population trapping and constructive contributions in resonantly enhanced nonlinear-optical frequency conversions and, at the same time, to distractive contributions in absorption of the fundamental and generated radiations^{1,2}. Wide range of applications are expected³.

Resonant nonlinear optical interference effects have been subject of the extensive both theoretical and experimental studies since the discovery of masers and lasers (see for example² and ref. therein). In this paper we briefly review some early and recent results of Russian research groups on this topic.

3 Resonant nonlinear optical interference processes**3.1 Destructive and constructive interference in classical and quantum optical physics**

Interference is one of the fundamental physical phenomena. Two oscillations at one and the same, or close, frequencies may interfere both in constructive and destructive ways. One can manipulate

¹Invited review paper

by the resulting oscillations with variation of the relative phase and the amplitudes of the interfering oscillators in order to enhance or, on the contrary, to eliminate the oscillations of any nature. Interference is widely used in optical physics, including quantum optics. The concept of interference is more general, then the notions of elementary quantum-optical processes, such as one-photon, multistep and multiphoton transitions. These notions were introduced and classify at their frequency-correlation properties in the framework of the perturbation theory. Indeed, in resonant interactions, these properties may be drastically changed with growth of the intensity of the coupled fields ^{4;2b,c}. The latter may give rise to such effects in nonlinear spectroscopy of Doppler broadened transition, as compensation of the residual inhomogeneous broadening in Raman-like and cascade configurations ^{4;2b,c;5}.

Quantum interference may occur when coherent superposition of real states is involved in a process⁶. Alternatively, interfering frequency-degenerate intraatomic oscillations may originate from different correlated quantum pathways, contributing in one and the same frequency. For example, in the weak-field approximation, these can be one- and two-photon contributions to an optical process, associated with the radiation at a given frequency. Such process may be thought as that started from the coherent superposition of closely spaced real energy-level and quasi-level (virtual state), created by the auxiliary strong field ^{2b,c;4}. Such a coherent superposition can be produced even more easily than in the case of real doublet state. In general, even in the cases, when many elementary processes contribute to an optical process and their classification is troublesome, one can explain and predict experimental results with the aid of the notion of interfering frequency-degenerated components of nonlinear polarization. The amplitudes of the components can be varied with the intensities and phases – with the frequency-detunings of the driving fields.

3.2 Effect of energy levels population and relaxation, density matrix approach

In general case of open energy-level configuration with all the levels being populated and various relaxation processes involved, density-matrix method is the most convenient for the analysis of a resonant nonlinear-optical response. Explicit formulae, describing spectral properties of a weak probe field in the presence of an auxiliary strong one, in cascade, V and Λ configurations can be easily derived in the similar way ^{2b,c}. We shall show that on the example of the energy-level schematic, given on Fig.1.

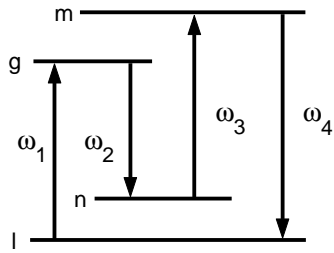


Figure 1: Atomic energy levels configuration.

Fields E_1 at frequency $\omega_1 \approx \omega_{gl}$ and E_3 at frequency $\omega_3 \approx \omega_{mn}$ are strong. Fields E_2 at frequency ω_2 and E_4 at frequency ω_4 are probe ones. We shall derive the conditions to achieve AWI at the transition gn , as well as at transition ml , so that both V and Λ configurations are embedded. Frequency of the probe field may be both higher and lower compared to the driving field.

Consider energy-level configuration, shown in the Fig.1. Density matrix equations in the interaction representation, relevant to the problem under consideration, can be written in the form:

$$\begin{aligned} \rho_{lg} &= r_1 \cdot \exp(i\Omega_1 t), \quad \rho_{nm} = r_3 \cdot \exp(i\Omega_3 t), \quad \rho_{ng} = r_2 \cdot \exp(i\Omega_1 t) + \tilde{r}_2 \cdot \exp[i(\Omega_1 + \Omega_3 - \Omega_4)t], \\ \rho_{lm} &= r_4 \cdot \exp(i\Omega_4 t) + \tilde{r}_4 \cdot \exp[i(\Omega_1 - \Omega_2 + \Omega_3)t], \quad \rho_{ln} = r_{12} \cdot \exp[i(\Omega_1 - \Omega_2)t] + r_{43} \cdot \exp[i(\Omega_4 - \Omega_3)t], \\ \rho_{ii} &= r_i, \\ P_2 r_2 &= iG_2 \Delta r_2 - iG_3 r_{32}^* + ir_{12}^* G_1, \quad d_2 \tilde{r}_2 = -iG_3 r_{41}^* + ir_{43}^* G_1, \\ P_4 r_4 &= i[G_4 \Delta r_4 - G_1 r_{41} + r_{43} G_3], \quad d_4 \tilde{r}_4 = -iG_1 r_{32} + ir_{12} G_3 \\ P_{41} r_{41} &= -iG_1^* r_4 + ir_1^* G_4, \quad P_{43} r_{43} = -iG_4 r_3^* + ir_4 G_3^*, \\ P_{32} r_{32} &= -iG_2^* r_3 + ir_2^* G_3, \quad P_{12} r_{12} = -iG_1 r_2^* + ir_1 G_2^*, \\ \Gamma_m r_m &= -2\text{Re}\{iG_3^* r_3\} + q_m, \quad \Gamma_n r_n = -2\text{Re}\{iG_3^* r_3\} + \gamma_{gn} r_g + \gamma_{mn} r_m + q_n, \\ \Gamma_g r_g &= -2\text{Re}\{iG_1^* r_1\} + q_g, \quad \Gamma_l r_l = -2\text{Re}\{iG_1^* r_1\} + \gamma_{gl} r_g + \gamma_{ml} r_m + q_l, \end{aligned}$$

$$\Delta r_1 = r_l - r_g, \quad \Delta r_2 = r_n - r_g, \quad \Delta r_3 = r_n - r_m, \quad \Delta r_4 = r_l - r_m.$$

Where $\Omega_1 = \omega_1 - \omega_{lg}$, $\Omega_3 = \omega_3 - \omega_{mn}$, $\Omega_2 = \omega_2 - \omega_{gn}$, $\Omega_4 = \omega_4 - \omega_{ml}$,
 $G_1 = -\mathbf{E}_1 \mathbf{d}_{lg}/2\hbar$, $G_2 = -\mathbf{E}_2 \mathbf{d}_{gn}/2\hbar$, $G_3 = -\mathbf{E}_3 \mathbf{d}_{nm}/2\hbar$, $G_4 = -\mathbf{E}_4 \mathbf{d}_{ml}/2\hbar$,
 $P_1 = \Gamma_{lg} + i\Omega_1$, $P_2 = \Gamma_{ng} + i\Omega_2$, $P_3 = \Gamma_{nm} + i\Omega_3$, $P_4 = \Gamma_{lm} + i\Omega_4$, $P_{12} = \Gamma_{ln} + i(\Omega_1 - \Omega_2)$,
 $P_{43} = \Gamma_{ln} + i(\Omega_4 - \Omega_3)$, $P_{32} = \Gamma_{gm} + i(\Omega_3 - \Omega_2)$, $P_{41} = \Gamma_{gm} + i(\Omega_4 - \Omega_1)$,
 $d_2 = \Gamma_{ng} + i(\Omega_1 + \Omega_3 - \Omega_4)$, $d_4 = \Gamma_{lm} + i(\Omega_1 - \Omega_2 + \Omega_3)$.

Here Ω_i are frequency detuning from the resonances, G_i — Rabi frequencies, Δr_i — power-depending population differences, Γ_{ij} — homogeneous half linewidths, Γ_i^{-1} — lifetimes, γ_{ij} — relaxation rates from i to j states, g_i — population rate by a incoherent source. Density matrix amplitudes r_i determine absorption/gain and refraction indexes, \tilde{r}_i — determine four – wave mixing driving nonlinear polarizations.

The equations and their solution for the cascade atomic configurations can be derived by the simple change of the detunings signs ^{2b}.

3.3 Laser-induced atomic coherence and classification of resonant nonlinear effects

Solution of the coupled density – matrix equations may be represented in the form:

$$r_{1,3} = iG_{1,3}\Delta r_1/P_1, \quad r_{2,4} = iG_{2,4}R_{2,4}/P_{2,4},$$

$$R_2 = \frac{\Delta r_2(1 + g_7 + v_7) - v_3(1 + v_7 - g_8)\Delta r_3 - g_3(1 + g_7 - v_8)\Delta r_1}{(1 + g_2 + v_2) + [g_7 + g_2(g_7 - v_8) + v_7 + v_2(v_7 - g_8)]}, \quad (1)$$

$$R_4 = \frac{\Delta r_4(1 + v_5 + g_5) - g_1(1 + g_5 - v_6)\Delta r_1 - v_1(1 + v_5 - g_6)\Delta r_3}{(1 + g_4 + v_4) + [v_5 + v_4(v_5 - g_6) + g_5 + g_4(g_5 - v_6)]}, \quad (2)$$

$$\Delta r_1 = \frac{(1 + \alpha_3)\Delta n_1 + b_1\alpha_3\Delta n_3}{(1 + \alpha_1)(1 + \alpha_3) - a_1\alpha_1b_1\alpha_3}, \quad \Delta r_3 = \frac{(1 + \alpha_1)\Delta n_3 + a_1\alpha_1\Delta n_1}{(1 + \alpha_1)(1 + \alpha_3) - a_1\alpha_1b_1\alpha_3},$$

$$\Delta r_2 = \Delta n_2 - b_2\alpha_3\Delta r_3 - a_2\alpha_1\Delta r_1, \quad \Delta r_4 = \Delta n_4 - a_3\alpha_1\Delta r_1 - b_3\alpha_3\Delta r_3;$$

$$r_m = n_m + (1 - b_2)\alpha_3\Delta r_3, \quad r_g = n_g + (1 - a_3)\alpha_1\Delta r_1, \quad r_n = n_n - b_2\alpha_3\Delta r_3 + a_1\alpha_1\Delta r_1, \quad (3)$$

$$r_l = n_l - b_1\alpha_3\Delta r_3 + a_3\alpha_1\Delta r_1, \quad \Delta r_i(E_1 = 0, E_3 = 0) = \Delta n_i;$$

$$g_1 = \frac{|G_1|^2}{P_{41}P_1^*}, g_2 = \frac{|G_1|^2}{P_{12}^*P_2}, g_3 = \frac{|G_1|^2}{P_{12}^*P_1^*}, g_4 = \frac{|G_1|^2}{P_{41}P_4}, g_5 = \frac{|G_1|^2}{P_{43}d_2^*}, g_6 = \frac{|G_1|^2}{P_{41}d_2^*}, g_7 = \frac{|G_1|^2}{P_{32}^*d_4^*}, g_8 = \frac{|G_1|^2}{P_{12}^*d_4^*},$$

$$v_1 = \frac{|G_3|^2}{P_{43}P_3^*}, v_2 = \frac{|G_3|^2}{P_{32}^*P_2}, v_3 = \frac{|G_3|^2}{P_{32}^*P_3^*}, v_4 = \frac{|G_3|^2}{P_{43}P_4}, v_5 = \frac{|G_3|^2}{P_{41}d_2^*}, v_6 = \frac{|G_3|^2}{P_{43}d_2^*}, v_7 = \frac{|G_3|^2}{P_{12}^*d_4^*}, v_8 = \frac{|G_3|^2}{P_{32}^*d_4^*};$$

$$\alpha_1 = \alpha_1^0 \frac{\Gamma_{lg}^2}{|P_1|^2}, \alpha_1^0 = \frac{2(\Gamma_l + \Gamma_g - \gamma_{gl})}{\Gamma_l \Gamma_g \Gamma_{lg}} |G_1|^2, \alpha_3 = \alpha_3^0 \frac{\Gamma_{mn}^2}{|P_3|^2}, \alpha_3^0 = \frac{2(\Gamma_m + \Gamma_n - \gamma_{mn})}{\Gamma_m \Gamma_n \Gamma_{mn}} |G_3|^2;$$

$$a_1 = \frac{\gamma_{gn}a_2}{\Gamma_n - \gamma_{gn}} = \frac{\gamma_{gn}\Gamma_l a_3}{\Gamma_n(\Gamma_g - \gamma_{gl})} = \frac{\gamma_{gn}\Gamma_l}{\Gamma_n(\Gamma_l + \Gamma_g - \gamma_{gl})},$$

$$b_1 = \frac{\gamma_{ml}\Gamma_n b_2}{\Gamma_l(\Gamma_m - \gamma_{mn})} = \frac{\gamma_{ml}b_3}{\Gamma_l(\Gamma_l - \gamma_{ml})} = \frac{\gamma_{ml}\Gamma_n}{\Gamma_l(\Gamma_m + \Gamma_n - \gamma_{mn})}.$$

By substituting frequency deviations Ω_i for that Doppler-shifted $\Omega_i - k_i v$ (v is atomic velocity) we can take into account the effect of atomic motion. Imaginary part of density-matrix amplitudes r_2 and r_4 represent absorption or gain at the corresponding probe-field frequencies. At $G_3 = 0$ equations (2) and (3) convert in solutions for Λ and V schemes

$$r_2 = i \frac{G_2}{P_2} \cdot \frac{\Delta r_2 - g_3 \Delta r_1}{1 + g_2}, \quad r_4 = i \frac{G_4}{P_4} \cdot \frac{\Delta r_4 - g_1 \Delta r_1}{1 + g_4}. \quad (4)$$

Following^{2b,c} we can classify resonant nonlinear effects as 1) power saturation of the populations (eq. (4)); 2) strong-field induced splitting of the probe-field resonances, or ac Stark effect (denominators in eqs. (4)); and 3) nonlinear interference effect (*NIEF*) (second and third terms in the nominators of eqs. (2)).

4 Difference in absorption and emission spectra due to the nonlinear interference effects, amplification without inversion, resonance-enhanced refraction without absorption

Power of emitted or absorbed radiation, for example at the frequency ω_2 , which is proportional to $Re(-iG_2^*r_2)$, can be considered as a difference between pure emission (the term, proportional to r_g) and pure absorption (the rest terms in eqs.(2)). The difference in frequency-dependence of these terms, induced by the auxiliary driving field, is the origin of *AWI*^{2b}. Refractive index at ω_2 is determined by $Im(-iG_2^*r_2)$ and, in general, laser-induced minimum in absorption may coincide with the resonance-enhanced maximum in refraction^{1,3}. Thus, *laser-induced resonance splitting and NIEF transform only spectral shape of absorption/gain and refractive indices, give rise to difference in the line shapes of spontaneous (or pure induced) emission and absorption, but do not affect the integral intensity of the spectral lines*^{2b,c}:

$$\int d\Omega_2 Re(-ir_2/G_2) = \Delta r_2, \quad \int d\Omega_4 Re(-ir_4/G_4) = \Delta r_4. \quad (5)$$

Indeed, *NIEF* give rise to electromagnetically induced transparency (*EIT*) and to *AWI* at the transitions gn (or ml), when contributions of second and third terms in the nominators of eqs.(2) are equal or dominate over Δr_2 (or over Δr_4), correspondingly. From the above presented density-matrix equations one can see that the coherence at the transitions gm and ln (r_{32} and r_{12}), induced in cooperation of the strong and the probe fields, is the source of the *EIT* and *AWI* effects.

A great number of elementary processes, introduced and defined for the bare states in the framework of the perturbation theory, may give contribution to the absorption/gain index $\alpha(\Omega_i)$. Consider, for example, $\alpha(\Omega_4)$ at the frequency $\omega_4 > \omega_1$ (Fig.1), reduced by it's maximum value $\alpha^0(0)$ in the absence of the all strong fields, for the case when $E_3 = 0$. From the eqs.(2) one finds:

$$\frac{\alpha(\Omega_4)}{\alpha^0(0)} = Re\left\{\frac{\Gamma_4}{P_4} \cdot \frac{\Delta r_4 - g_1 \Delta r_1}{\Delta n_4(1 + g_4)}\right\} \quad (6)$$

Consider two subcases:

a. Off resonance: $|\Omega_1| \approx |\Omega_4| \gg \Gamma_1, \Gamma_4; |g_4| \ll 1; |g_1| \ll 1; P_4 \approx i\Omega_4; P_1 \approx i\Omega_1 \approx i\Omega_4$.

Eq.(6) takes the form:

$$\begin{aligned} \frac{\alpha(\Omega_4)}{\alpha^0(0)} &\approx \frac{\Gamma_4^2 \Delta r_4}{\Omega_4^2 \Delta n_4} - Re\left\{\frac{\Gamma_4(\Delta r_4 g_4 + \Delta r_1 g_1)}{i\Omega_4 \Delta n_4}\right\} \approx \frac{\Gamma_4^2 \Delta r_4}{\Omega_4^2 \Delta n_4} - \frac{\Gamma_4 \Gamma_{14}}{\Gamma_{14}^2 + (\Omega_4 - \Omega_1)^2} \cdot \frac{|G_1|^2 (\Delta r_1 - \Delta r_4)}{\Omega_4^2 \Delta n_4} = \\ &= \frac{\Gamma_{lm}^2 (r_l - r_m)}{(n_l - n_m) \Omega_4^2} - \frac{\Gamma_{gm} \Gamma_{lm}}{\Gamma_{gm}^2 + (\Omega_4 - \Omega_1)^2} \cdot \frac{|G_1|^2 (r_m - r_g)}{\Omega_4^2 (n_l - n_m)} \end{aligned} \quad (7)$$

The last terms in eqs.(7) describe Raman-like coupling and originate both from the nominator and the denominator in eq.(6). *Population inversion between initial and final bare states ($r_m = n_m > r_g$) is required for amplification of the probe field.*

b. Resonance: $\Omega_1 = \Omega_4 = 0$.

Conditions for *AWI* and *EIT* are:

$$g_1 \Delta r_1 \geq \Delta r_4, \quad \text{or} \quad \frac{|G_1|^2}{\Gamma_l \Gamma_{gm}} \cdot (r_l - r_g) \geq r_l - r_m \quad (8)$$

Eq.(8) shows that *due to NIEF, population inversion between initial and final bare states is not required in order to attain AWI in this case. Small relaxation rate of the coherence, induced in cooperation of the driving and probe fields, compared to the other relaxation rates is the most important.*

Analysis of the condition for *EIT* and *AWI* as well as of sign-changing line shape in the more details can be found in ref.^{2b} both for open and closed (l is ground state) atomic configurations. The analysis shows strong dependence of the line shape on the ratios of both population and coherence relaxation rates as well as on the ratios of initial unsaturated population differences on the coupled transitions.

4.1 Constructive and destructive interference due to the atomic velocity distribution

Furthermore, the analysis shows that the contributions of the coherence driving fields to the spectra may be both constructive and destructive, depending on the detunings of the probe as well as of the strong fields. This indicates that in gases with inhomogeneous broadening of the coupled transitions, dominating over homogeneous one, conditions for *AWI* and *EIT* may considerably differ from that for atoms at rest. Nevertheless, it was found out that under certain conditions sign-changing spectral profiles may be produced too^{2b,c;7a,b}. At weak intensities of driving field narrow structure, superimposed on the Doppler background, appears. The shape of the structure is anisotropic and depends on the angle between the wave vectors of the interacting radiations. Optically-pumped unidirectional-emitting ring laser may operate by that. The line shape is also dependent on the intensity of the driving field and velocity-changing collisions. Special features may occur, when some of the coupled transitions are homogeneously, and some of them are inhomogeneously broadened. It was found out that destructive or constructive character of the effect of Maxwell's velocity distribution depends on the fact whether a frequency of the probe field is less or greater than that of the strong one too. Analytical results describing general behavior of the velocity-averaged functions for some limiting cases, including Rabi frequencies larger than homogeneous linewidths, can be found in ref.^{2a,b,c;7a,b}.

5 Coherence and nonlinear-optical conversion, Enhancements in nonlinear-optical conversion due to multiple resonance and induced transparency, Local-field effects

Nonlinear optical response of a medium experiences a giant enhancements in one- and multiphoton resonances. This reduces required fundamental powers down to cw regime⁸, however imposes severe limitations on the number density of the medium due to absorption of fundamental and generated radiations. As it is discussed above, in the presence of a strong electromagnetic radiation resonances for a weak probe radiation experience splitting^{2,9}, which exhibits itself in a different ways in real and imaginary parts of linear and nonlinear susceptibilities. Later makes possible to *combine decrease in absorption with increase in squared module of nonlinear susceptibilities*, responsible for optical generation, and at the same time with *improvements in phase-matching and increasing density of the medium*^{2f,10}.

With the increase of the atom number density, *local field*, acting on an atom, may pretty much differ from the external field both in the amplitude and phase. This *may drastically change shape of nonlinear spectroscopic structures, including electromagnetically induced transparency*^{11,12}.

Consider experimental schematics, proposed in ref.¹³, that combines the advantages of both *multiple resonance enhancements and increase in atom number density of nonlinear medium* due to the above

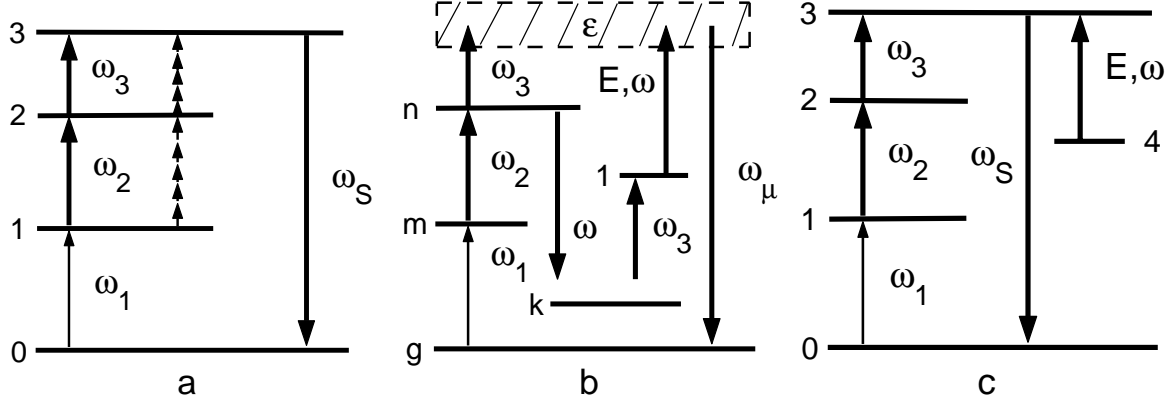


Figure 2: Interference enhanced frequency-mixing. *a.* Triple resonance enhanced frequency-conversion. Levels 2 and 1 as well as 3 and 2 are coupled by the strong either one- or multiphoton interactions. Levels 1 and 2 are coupled by the weak field. *b.* Enhancement in frequency-mixing due to the autoionizing-like resonance, induced in spectral continuum by the auxiliary strong fields E and E_3 . *c.* Enhancement in three-photon resonant four-wave mixing, induced by the auxiliary strong field E .

mentioned Autler-Townes (*ac* Stark splitting) as well as *local-field effects*.

Consider energy-level scheme, shown in Fig.2a. Strong fields at frequencies ω_3 and ω_2 couple unpopulated levels 3 and 2 (Rabi frequency G_3) and 2 and 1 (Rabi frequency G_2), respectively. Field at $\omega_1 \approx \omega_{10}$ as well as generated $\omega_s = \omega_1 + \omega_2 + \omega_3$ are weak, do not change populations of the levels and are accounted for only in the lowest order of the perturbation theory. Absorption and refraction indexes for the probe fields at ω_1 and ω_s are represented by the imaginary and real parts of

$$\chi_1(-\omega_1; \omega_1) = (\chi_1^0/P_{01})f_1, \quad \chi_s(-\omega_s; \omega_s) = (\chi_s^0/P_{03})f_s \quad (9)$$

Nonlinear susceptibility is:

$$\chi^{NL}(-\omega_s; \omega_1 + \omega_2 + \omega_3) = (\chi_0^{NL}/P_{01}P_{02}D_{03})f, \quad (10)$$

where χ_1^0, χ_s^0 and χ_0^{NL} are resonant values of the susceptibilities at negligibly small G_2 and G_3 . Factors f_1, f_2 and f describe effects of the strong fields. Simple density - matrix calculations, similar to given in^{2b,e;10a}. yield:

$$f_1 = \{1 + g_2/P_{01}P_{02}[1 + (g_3/P_{02}D_{03})]\}^{-1}, \quad (11)$$

$$f_s = \{1 + g_3/P_{03}D_{02}[1 + (g_2/D_{02}D_{01})]\}^{-1}, \quad (12)$$

$$f = f_1[1 + g_3/D_{03}P_{02}]^{-1} = [1 + (g_2/D_{02}D_{01}) + (g_3/D_{03}P_{02})]^{-1} \quad (13)$$

$$P_{01} = 1 + ix_1, \quad P_{02} = 1 + ix_0, \quad P_{03} = 1 + ix_s; \quad D_{01} = 1 + iy_1, \quad D_{02} = 1 + iy_0, \quad D_{03} = 1 + iy_s;$$

$$x_1 = (\omega_1 - \omega_{10})/\Gamma_{10} = 0, \quad x_{02} = (\omega_1 + \omega_2 - \omega_{21})/\Gamma_{20} = 0, \quad x_s = (\omega_s - \omega_{30})/\Gamma_{30} = 0;$$

$$y_1 = (\omega_s - \omega_3 - \omega_2 - \omega_{10})/\Gamma_{10} = 0, \quad y_{02} = (\omega_s - \omega_3 - \omega_{21})/\Gamma_{20} = 0, \quad y_s = (\omega_1 + \omega_2 + \omega_3 - \omega_{30})/\Gamma_{30} = 0;$$

$$g_2 = G_2^2/\Gamma_{10}\Gamma_2, \quad g_3 = G_3^2/\Gamma_{30}\Gamma_{20},$$

Γ_{ij} are homogeneous halfwidth of the corresponding transitions. In the case, when E_s is not a probe field, but generated radiation, $\omega_s = \omega_1 + \omega_2 + \omega_3$ and $D_{0i} = P_{0i}$.

Factors f_1, f_s and f are different and describe splitting of the corresponding resonances. Frequency-dependence and difference from unity of the factors f_1, f_s and f is determined by the coherence, induced at the transition 02 by the two combinations of strong and weak fields (E_1, E_2 and E_s, E_3).

Generated power $P \propto g_2 g_3 |\chi^{NL}|^2$, depends not only on imaginary but on real part of χ^{NL} too, and because of that may not deplete in the spectral range of induced transparency and phase-matching. Each resonance increases $|\chi^{NL}|^2$ by the factor of x_i^{-2} , which may be on the order of 10^6 . Laser-induced spectral structures in real parts of χ_1 and χ_s (dispersion caused by the coherence at the 02 transition), provide additional means to phase-match frequency - conversion by the small detunings of the fundamental radiations from the resonances. Triple resonance may yield total enhancement in generated power on the order of 10^{18} . Due to the induced transparency, number density of the atoms N and consequently $P \propto N^2$ may be increased by several orders of the magnitude in addition.

At high number density of the atoms, local fields may significantly differ from the external electromagnetic fields both in amplitudes and in phases. As it was shown in ^{12,13}, that may drastically change spectral properties of the induced transparency as well as of the generating nonlinear polarization. Similar to ^{11,12}, making use Lorentz-Lorenz approximation, local field effects can be accounted for by the substituting one-photon resonances on that red-shifted (by substituting x_1 and x_s for $x_1 + C_1$ and $x_s + C_s$, $C_1 = N |d_{10}|^2 / 3\epsilon_0 \Gamma_{10}$; $C_s = N |d_{30}|^2 / 3\epsilon_0 \Gamma_{30}$, ϵ_0 - is permittivity of free space). Due to the fact that this does not influence transition frequencies between the excited states and that of the multiphoton transitions, the introduced shifts may drastically change effects of strong electromagnetic fields at ω_2 and ω_3 on both dressed linear and nonlinear responses.

Equations, given above, can be easily generalized on the cases of the *higher order processes*. For example, when 1-0 and/or 3-2, 2-1 are multiphoton transitions, generalization can be done simply by substituting one-photon Rabi frequencies and detunings for the corresponding multiphoton ones. Manipulations by the nonlinear susceptibility, absorption and refractive indexes for the generating radiation with the *auxilliary* strong fields, coupled to the *adjacent* transitions (both bound and continuum states, Figs. 2b, c.), were considered in ref.^{2f,10}.

6 Nonlinear interference effects at bound-free transitions, Laser-induced autoionizing-like resonances (laser induced continuum structure)

Nonlinear interference phenomena, similar to those at bound-bound transitions, including *AWI* and *EIT*, can occur at the transitions to ionization continuum. Appropriate theory was developed in ref. ^{2f,10a,14}. Similar case, relevant to the zone bands in crystals, was considered in ref.¹⁵. Laser induced autoionizing like resonances – laser induced continuum structure (*LICS*) was observed in the experiments ref.¹⁶, and since the end of 80's studies of the resonant interference processes in the context of *LICS*, *AWI* and *EIT*, first at bound-free and then at bound-bound transitions, have involved a number of research groups^{17,18}.

Potential feasibilities to manipulate both by *LICS* and by the splitting of the discrete resonances in order to enhance short - wavelengths frequency - mixing output and to decrease resonant absorption of the both fundamental and generated radiations can be shown with the example of Fig.2b., generalized for the case, when ω_1 is close to ω_{10} , and radiations at ω_2 , ω_3 and ω are strong. The example combines opportunities to manipulate by two *LICS* and by depletion of absorption at the discrete transitions. Contribution of strong off - resonant k levels are taken into account too. By that, the detunings $|\omega_1 - \omega_{gm}|$, $|\omega_1 + \omega_2 - \omega_{gn}|$ and $|\omega - \omega_3 - \omega_{nl}|$ are assumed being much less than all the rest. Density - matrix calculations give the expressions for nonlinear susceptibility $\chi^{(3)}(\omega_\mu = \omega_1 + \omega_2 + \omega_3)$, which determines generated power at the frequency ω_μ , as well as for absorption indexes $\alpha(\omega_1)$ and $\alpha(\omega_\mu)$ for probe radiations at corresponding frequencies as follows ¹⁹:

$$\chi^{(3)}(\omega_\mu = \omega_1 + \omega_2 + \omega_3) / \chi_{0\mu}^{(3)} = K / (D_{gm} X), \quad (14)$$

$$\alpha(\omega_1) / \alpha_{01} = \text{Re} \{ [1 - g_{mn} / (D_{gm} X)] / D_{gm} \}, \quad (15)$$

$$\alpha(\omega_\mu)/\alpha_{0\mu} = 1 - k_3\beta_l + k_3\beta_l(y_l + q_{gl})^2/(1 + y_l^2) - \text{Re}\{k_4g_{nn}A^2(1 - iq_{gn})^2/Y\} \quad (16)$$

where $\chi_{0\mu}^{(3)}$, α_{01} and $\alpha_{0\mu}$ - are corresponding resonant values at the intensities of all the fields being negligibly weak. The rest parameters are as follows:

$$K = 1 - k_1\beta_l[(1 - iq_{nl})(1 - iq_{lg})]/[(1 - iq_{ng})(1 + ix_l)], \quad (17)$$

$$A = 1 - k_1\beta_l[(1 - iq_{ln})(1 - iq_{gl})]/[(1 - iq_{gn})(1 + iy_l)], \quad (18)$$

$$X = (1 + g_{nn})[1 + ix_n + g_{mn}/D_{gm}(1 + g_{nn}) - k_2\beta_l\beta_n(1 - iq_{nl})^2/(1 + ix_l)], \quad (19)$$

$$Y = (1 + g_{nn})[1 + iy_n + g_{mn}/p_{gm}(1 + g_{nn}) - k_2\beta_l\beta_n(1 - iq_{nl})^2/(1 + iy_l)], \quad (20)$$

$$D_{gm} = 1 + i(\omega_1 - \omega_{gm})/\Gamma_{gm}, \quad p_{gm} = 1 + i(\omega_\mu - \omega_3 - \omega_2 - \omega_{gm})/\Gamma_{gm}, \quad (21)$$

$$x_l = (\omega_1 + \omega_2 + \omega_3 - \omega - \omega_{gl} - \delta_{ll})/(\Gamma_{gl} + \gamma_{ll}), \quad x_n = (\omega_1 + \omega_2 - \omega_{gn} - \delta_{nn})/(\Gamma_{gn} + \gamma_{nn}), \quad (22)$$

$$y_l = (\omega_\mu - \omega - \omega_{gl} - \delta_{ll})/(\Gamma_{gl} + \gamma_{ll}), \quad y_n = (\omega_\mu - \omega_3 - \omega_{gn} - \delta_{nn})/(\Gamma_{gn} + \gamma_{nn}), \quad (23)$$

$$k_1 = (\gamma_{gl}\gamma_{ln})/(\gamma_{gn}\gamma_{nn}), k_2 = (\gamma_{nl}\gamma_{ln})/(\gamma_{ll}\gamma_{nn}), k_3 = (\gamma_{gl}\gamma_{lg})/(\gamma_{gg}\gamma_{ll}), k_4 = (\gamma_{gn}\gamma_{ng})/(\gamma_{gg}\gamma_{nn}), \quad (24)$$

$$g_{mn} = |G_{mn}|^2 / \Gamma_{gm}\Gamma_{gn}, \quad \beta_l = g_{ll}/(1 + g_{ll}), \quad \beta_n = g_{nn}/(1 + g_{nn}), \quad (25)$$

$$g_{ii} = \gamma_{ii}/\Gamma_{gi}, \quad q_{ij} = \delta_{ij}/\gamma_{ij}, \quad \gamma_{ij} = \pi\hbar G_{i\epsilon}G_{\epsilon j}|_{\epsilon=\hbar\omega_\mu} + \text{Re}\{\sum_k G_{ik}G_{kj}/p_{gk}\}, \quad (26)$$

$$\delta_{ij} = \hbar P \int d\epsilon G_{i\epsilon}G_{\epsilon j}/(\hbar\omega_\mu - \epsilon) + \text{Im}\{\sum_k G_{ik}G_{kj}/p_{gk}\} \quad (27)$$

Factors $0 \leq k_i \leq 1$, depending on whether continuum states are not degenerate or degenerate (unity).

Comparing eqs.(14) and (16) with corresponding equations from ref.^{10a,2f}, one can see additional interference *LICS* structures in generating nonlinear polarization, absorption and refraction indexes, produced in cooperation by the E_3 and E fields (terms, proportional to β_n and g_n), which provide with the supplementary means in absorption spectroscopy and for enhancements of generated short-wavelength radiation.

7 Relaxation-induced coherence processes

As it was outlined above, relaxation may influence interference processes both in negative and positive ways. Consider examples, when role of relaxation is positive.

7.1 *AWI* due to interference in spontaneous cascade of polarizations

The features in absorption and emission spectra, discussed above, are caused by interference of contributions of probe field and combination of probe and auxiliary strong field in atomic polarization. As it was outlined above, there may be other sources of interfering intraatomic oscillations. One of the means to obtain *AWI* without making use of auxiliary strong fields has been suggested recently in ref.²⁰. The origin is interference through the correlations in spontaneous decay.

Consider four-level atomic configuration shown in Fig.3a. All four transitions are allowed. Suppose, that the transition frequency ω_{mn} is close to $\omega_{m_1n_1}$, and ω_{m_1m} is close to ω_{n_1n} , that is difference Δ

$$\Delta = \omega_{m_1n_1} - \omega_{mn} = \omega_{m_1m} - \omega_{n_1n} \quad (28)$$

is small. In this case interference between considered four radiating channels is possible. It is caused by the coherence transfer due to interaction with the vacuum oscillations, besides the populations

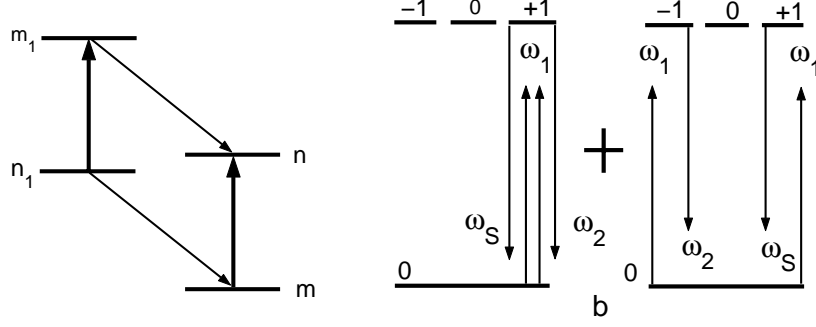


Figure 3: Energy-level schematics for relaxation-induced coherent processes. *a.* *AWI* through spontaneous cascade of polarizations. *b.* Relaxation-induced *FWM*.

decay and spontaneous emissions of photons. For the absorption index in the frequency range around ω_{mn} calculations give:

$$\alpha(\Omega) = \frac{\lambda^2}{4\pi} \{ N_{nm} A_{mn} \frac{\Gamma}{\Gamma^2 + \Omega^2} + N_{n_1 m_1} A_{m_1 n_1} [\frac{\Gamma_1}{\Gamma_1^2 + (\Omega - \Delta)^2} + \frac{KC}{\Gamma \Gamma_1} f(\Omega)] \}, \quad (29)$$

$$C = \sqrt{A_{m_1 m} A_{n_1 n} A_{mn} / A_{m_1 n_1}}, \quad K = (-1)^{J_m + J_{n_1}} \sqrt{2J_m + 1} \sqrt{2J_{n_1} + 1} \begin{Bmatrix} J_m & J_n & 1 \\ J_{n_1} & J_{m_1} & 1 \end{Bmatrix}, \quad (30)$$

$$f(\Omega) = \text{Re} \frac{\Gamma \Gamma_1}{(\Gamma - i\Omega)[\Gamma_1 - i(\Omega - \Delta)]} = \frac{\Gamma \Gamma_1 [\Gamma \Gamma_1 - \Omega(\Omega - \Delta)]}{(\Gamma^2 + \Omega^2)[\Gamma_1^2 + (\Omega - \Delta)^2]}, \quad (31)$$

$$N_{nm} = (2J_m + 1)(\rho_n - \rho_m), \quad N_{n_1 m_1} = (2J_{m_1} + 1)(\rho_{n_1} - \rho_{m_1}). \quad (32)$$

Here $\Omega = \omega - \omega_{mn}$, A_{ij} – Einstein coefficients, Γ, Γ_1 – are line halfwidths for the interfering transitions, J_i – energy level momenta, N_{ij} – population differences.

The interference term is described by the function $f(\Omega)$, $\int f(\Omega) d\Omega = 0$. Coefficient K is determined by the moments of four levels under consideration and may vary in the interval $-1 \leq K \leq 1$. The case $K \geq 0$ corresponds to constructive interference (enhancements in the oscillations), the case $K \leq 0$ — to destructive interference. The analysis of the lineshape eq.29 shows its sign-changing behavior. For example, at $|\Omega| \gg \Delta$

$$\alpha(\Omega) = \frac{\lambda^2}{4\pi\Omega^2} \{ N_{nm} A_{mn} \Gamma + N_{n_1 m_1} A_{m_1 n_1} (\Gamma_1 - KC) \}, \quad (33)$$

According to eq.33, absorption index may occur negative (*AWI*), if the requirements

$$K > 0, \quad (KC/\Gamma_1 - 1) N_{n_1 m_1} A_{m_1 n_1} \Gamma_1 > N_{nm} A_{mn} \Gamma \quad (34)$$

are met. When $K \leq 0, \Delta = 0$, the condition

$$(|K|C/\Gamma - 1) N_{n_1 m_1} A_{m_1 n_1} \Gamma > N_{nm} A_{mn} \Gamma_1 \quad (35)$$

means appearance of *AWI* in the line center ($\Omega = 0$). Similar phenomena may occur in the spectral range of the doublet $\omega_{m_1 m}, \omega_{n_1 n}$. Thus, in the considered atomic configuration *AWI* may be provided by the correlations in the spontaneous decay without any external action.

7.2 Collision-induced four-wave mixing

Consider example, when collisions and spontaneous relaxation, as well as external magnetic field, break destructive interference^{8a}. This remove elimination of for-wave mixing process and provides with the test, selectively sensitive to the specific modes of relaxation. The experiment was carried out with $He - Ne$ laser, $\lambda = 1.52\mu m$, which is resonant to $2s_2-2p_4$ transition of Ne . The upper level consist of three Zeeman's sublevel ($J_1 = 1$), the lower one is singlet ($J_0 = 0$). Fundamental beam consisted of two linear and orthogonal polarized components E_1 and E_2 , frequency-shift $\Delta = \omega_2 - \omega_1$ being much less than natural transition linewidth. Intensity of the radiation at ω_1 was much greater then that at ω_2 . Collision and magnetic field sensitive four-wave mixing output E_s at $\omega_s = 2\omega_1 - \omega_2 = \omega_2 - 2\Delta$ and with the same polarization as E_2 was detected. Growth of the *FWM* signal with the increase of collision rate and strength of magnetic field was observed, that can be explained as follows.

Each field and emitting nonlinear polarization $P^{NL}(\omega_s)$ may be represented as combination of two circular polarized components $P_+^{NL}(\omega_s)$ and $P_-^{NL}(\omega_s)$. Formulae for each of these components of non-linear polarization consist of two terms. One of them describes *FWM* of the radiations with one and the same polarizations in two-level subsystem, another one – *FWM* of the waves with opposite polarizations in three-level Zeeman's subsystem (Fig.3b). In the schematic under consideration, it turned out, that the two contributions interfere in the distractive way and completely eliminate each other, provided by the relaxation rates of population and quadruple moment (alignment) in the upper level are equal. It is obvious that trapping of the spontaneous radiation from the upper level, anisotropic collisions, as well as external magnetic field break the counterbalance and, therefore, induce *FWM* output. Such dependence was observed in the experiments. External magnetic field turns the second channel into fully resonant double-*V* schematics.

8 Review of early theory and experiments on *NIEF*, *AWI* and related phenomena

Coherence phenomena in three-level systems were studied since discovery of *masers*. Feasibility to attain *AWI* in these systems was discussed in some of publications of that period both for microwave²¹ and optical transitions²². *AWI* in optical two-level systems was predicted in ref.²³ and first was observed in radio-frequency transitions^{24,2d}. In optical range *AWI* and corresponding features in refractive index were observed in ref.^{2e,25}. Studies of coherence and interference phenomena in quantum transitions is growing research area, since they are embedded in many optical processes of basic and practical importance.

9 Concluding remarks

As it was outline, interference is basic and very general phenomenon of optical physics, which may play a crucial role in many experimental schematics of resonant nonlinear optics. Some of such schematics are shown in the Fig.4.

Fig.4a. shows upconversion of weak infrared radiation at the frequency ω_2 . Fields E_1 and E_3 are strong. Destructive interference of oscillations at the frequency $\omega_s - \omega_3 = \omega_{ng} = \omega_1 + \omega_2$ was shown to be one of the main process, limiting the conversion efficiency²⁶. Fig.4b. – interference of multiphoton transition and one-photon, induced by the generating radiation eliminates population of the upper level. Fig.4c. – off-resonant 7th-order seventh-harmonic generation interfere with resonant 9th-order seventh-harmonic generation, that was used for detection of the processes²⁶. Figs.4d,e. – interference of contributions of the doublet sublevels in two-photon and off-resonant one-photon transitions. Figs.4f,g. – interference of doublet sublevels in *FWM*.

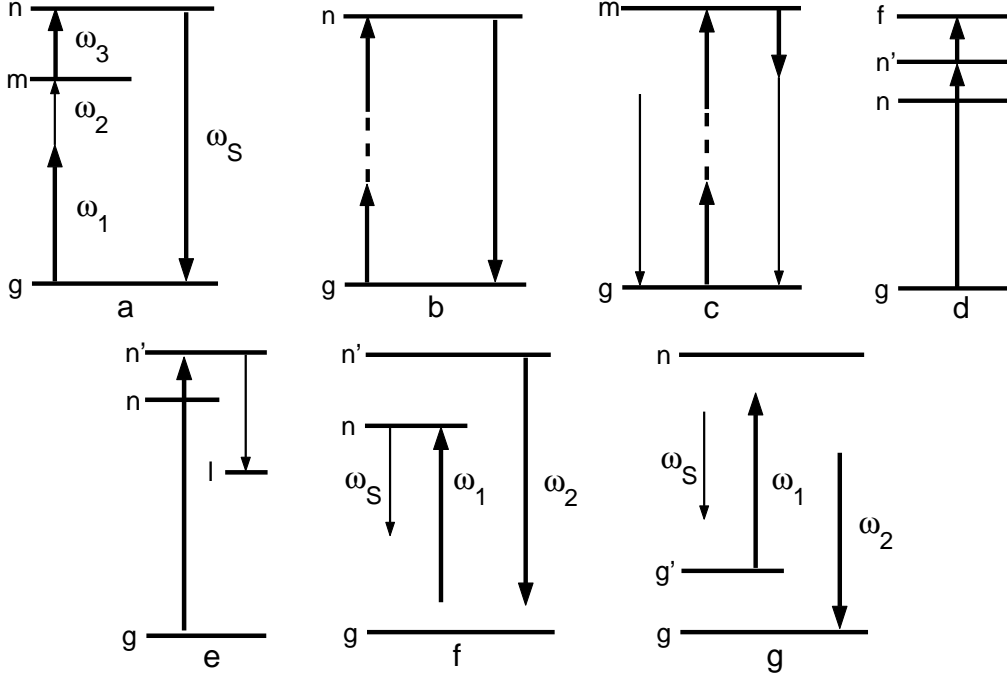


Figure 4: Interfering channels, embedded in resonant nonlinear optical processes.

Pressure-induced resonance was first proposed and experimentally proved in^{8a} and later in²⁷. *The entire analogy* between the schemes 3b and 4f is seen from the formula for the driving coherence (scheme 4f)

$$\begin{aligned} \rho_{n'n}^{(2)} &\propto V_{n'g}\rho_{gn}^{(1)} + \rho_{n'g}^{(1)}V_{gn} \propto \left[\frac{1}{\Omega_2 + i\Gamma_{n'g}} - \frac{1}{\Omega_1 - i\Gamma_{ng}} \right] \frac{1}{\Omega + i\Gamma_{n'n}} = \\ &= \frac{1}{(\Omega_2 + i\Gamma_{n'g})(\Omega_1 - i\Gamma_{ng})} \left[1 - i \frac{\Gamma_{nn'} - \Gamma_{n'g} - \Gamma_{ng}}{\Omega + i\Gamma_{nn'}} \right]. \end{aligned} \quad (36)$$

Here $\Omega_1 = \omega_1 - \omega_{ng}$, $\Omega_2 = \omega_2 - \omega_{n'g}$, $\Omega = \omega_2 - \omega_1 - \omega_{n'n}$. At spontaneous relaxation, $\Gamma_{ij} = (\Gamma_i + \Gamma_j)/2$, and resonance $\Omega = 0$ disappears. Collisions induce this resonance.

10 Acknowledgements

This work was supported in part by the International Science Foundation (Grants No 4000, No 4300), by the Russian Foundation for Fundamental Research (Grants N 93-02-03460, N 4300) and by the Krasnoyarsk Regional science Foundation, (Grant 4F 0123).

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